

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE



In re Application of	) Examiner: Thuan D. Dang
RONALD E. NICHOLS et al.	) Group Art Unit 1764
Serial No.: 09/925,391	) Entitled:
Filed: August 9, 2001	) LOW ENERGY METHOD OF
Atty Docket No.: 287122-00004-2	) PYROLYSIS OF HYDROCARBON
	) MATERIALS SUCH AS RUBBER

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DECLARATION OF ALAN M. LEVINE

I, Alan M. Levine, being duly sworn hereby declare as follows:

1. I have a B.S. and M.S. degree in physics from Stevens Institute of Technology, Hoboken, New Jersey, awarded in 1971 and 1973, respectively, and a Ph.D. from the University of Pittsburgh, Pittsburgh Pennsylvania, awarded in 1979.

2. From 1992 to the present I have been employed at RJ Lee Group, Monroeville, PA, as manager of the organic and inorganic chemistry laboratories. As manager, I identify new markets for organic and inorganic chemistry and X-ray diffraction laboratories, have day-to-day supervision of a chemistry department consisting of 12 employees in 9 laboratories, and provide oversight of routine sample analyses and quality assurance of procedures and results. I am also a senior technical consultant at RJ Lee Group, and am responsible for technical support and direction of a pyrolysis process research and development program.

3. My professional experience prior to 1992 is listed in Appendix A. I have over 25 years of laboratory science experience, in the biological and chemical sciences.

4. I am an author or coauthor of the publications and presentations listed in the attached Appendix B.

5. I am a named inventor on the present application. I participated in the preparation of the patent application, and carefully reviewed the application prior to its filing. I have read the previous Office Action issued in this case, and participated in the response to that Office Action. I have read the outstanding Office Action and the references cited by the Examiner, and have participated in the response to the Office Action, including responding to the rejection made under 35 USC §103.

6. The Gi reference is cited as teaching the use of metals and clay as a catalyst in a method of pyrolysis. According to the Examiner, it would have been obvious

to combine the teachings of Gi (4463203) with the teachings of Roy (4740270) or Solbakken (4250158), both said to teach methods of low pressure pyrolysis, to arrive at the present invention. It is my well considered opinion that this assertion is incorrect.

7. The Gi patent does not teach the use of metals in elemental form as a catalyst in the pyrolysis process. In fact, Gi does not teach the use of clay as a catalyst at all; brown coal is said to act as a catalyst at column 2 line 68. Gi discloses that clay prevents sticking of the hydrocarbon material to the reactor chamber at column 3, 21-23. In addition, the temperatures claimed by Gi would be too high to take advantage of any catalytic effects since they are well above the energy needed for pyrolytic cracking and at temperatures that inactivate bentonite's catalytic effects by collapsing the layers of the clay. Thus, one skilled in the art would receive no guidance on the selection of clay catalysts from the Gi reference.

8. It is said to be obvious to combine the teachings of Gi with Solbakken, to arrive at the present invention. As recognized by the Examiner, Solbakken teaches a pyrolysis method in which low pressure is used to optimize the oil yield, not the carbonaceous product. Solbakken does not teach the use of a catalyst, and uses much higher temperatures than those used in the present invention: 750°-1800°F, as compared with 450°-850° in the present invention. Solbakken does not disclose that use of a low pressure system in combination with the temperatures used in the present invention would provide complete pyrolysis and an improved carbon black product. One skilled in the art would find no guidance in Solbakken on how to use temperatures lower than those disclosed, and yet achieve an improved carbon black product. Generally, one assumes that lowering temperatures in pyrolysis will result in incomplete pyrolysis of the hydrocarbon material, and in the absence of specific guidance one skilled in the art would not expect that lower temperatures would suffice.

9. Similarly, it is said to be obvious to combine the teachings of Gi with Roy. Roy does not teach a catalyst, and also attempts to reduce the amount of carbon black product produced. The low pressure used by Roy is insignificant, compared with that used in the present invention (<1.38 inches Hg in Roy compared to 2-16 inches Hg in the present invention). One skilled in the art finds no guidance in Roy regarding the use of lower pressures, and no guidance on selecting optimal pressures and temperatures for improving the carbon black product.

10. The Gi reference is also cited as disclosing the use of at least three temperature zones in the pyrolysis process. It is my well considered opinion that this is an incorrect reading of the Gi reference. Gi merely shows how the reaction proceeds at the

various temperatures; it does not disclose a method in which affirmative steps are taken to reduce the energy input in different phases, to take advantage of the energy produced by the pyrolysis process.

11. Based on the above, it would not have been obvious to combine the teachings of the references cited to arrive at the present invention. This conclusion is further supported by the extensive testing conducted by me and others at RJ Lee Group, Inc., in which we have optimized the pyrolysis process to provide an economical and environmentally sound method of producing high quality carbon black.

12. We conducted initial testing in several batch bench reactors that were designed and built in-house. These units are constructed of 20 quart metal reaction vessels attached to condensing coils and vacuum pumps and are heated with 180,000 BTU propane burners. These systems are designed to pyrolyze approximately 2 lbs of hydrocarbon material (e.g. rubber). Tests in the bench reactors have supported the commercial viability of pyrolysis of rubber and other hydrocarbons and the effectiveness of various catalyst formulations in lowering the initiation temperature of the reaction and the process energy requirement. The tests were typically performed under the following conditions. Catalyst is mixed in the reaction vessel with rubber chips at a rate of five grams per pound of rubber. The system is sealed and the vacuum is brought to 10 "Hg. Starting at ambient temperature, we heat with propane burners set to approximately 30 lpm propane flow until the condensed oil drips steadily at approximately 1 drop per second. 10"Hg of vacuum are maintained throughout the heating and cool down. At that point the gas is turned off and the vacuum is maintained while we continued to monitor the temperature. The vacuum is removed after the reaction product cools to below 130°F and the product is completed. The complete process typically is completed in 2.5 to 3 hours. Our focus for this testing has been to produce high quality carbon black economically and in an environmentally conscious manner.

13. Carbon black derived from the bench reactor studies was analyzed. Two classes of potential carbon black contaminants were identified, an oil-like material that boils off below 752°F and a pitch-like material that requires approximately 790°F to boil off. This is consistent with the "carbonaceous deposits" that Darmstadt describes in several publications as interfering with binding of rubber to carbon black and is more effectively removed during pyrolysis as temperatures and vacuums are increased. We isolated this material by hot solvent extractions and demonstrated that bentonite breaks some of it down at temperatures below 250°F. Thus, the use of bentonite alone or bentonite in combination with metals effects the removal of this pitch-like residue during pyrolysis.

14. Side-by-side bench runs of hydrocarbon material with and without various catalyst formulations at the same heat inputs have demonstrated the following:

- (i) The catalyzed reaction with bentonite alone or bentonite in combination with metals progresses at a higher temperature starting between 100°F and 150°F (FIGURE 1).
- (ii) When heat is removed during the reaction, temperatures continue to rise with bentonite and bentonite/metals catalyzed reaction temperatures peaking higher than the non-catalyzed reaction temperatures (FIGURE 1).
- (iii) The increase in temperature is 20°F to 100°F when bentonite is used alone.
- (iv) The increase in temperature is 100° to 250°F when the bentonite /metals formulation is used.
- (v) The increase is 150°F to 300°F when the pillared bentonite formulation is used.

This testing demonstrates that the catalysts tested boost the reaction rate, with pillared bentonite being most effective, bentonite/metals next and bentonite alone next effective. Under the same energy inputs with either catalyst, oil and gas are produced earlier and in greater amounts than without catalyst and products from the catalyzed reactions contained significantly less unpyrolyzed material. This demonstrates that less energy is required to breakdown the feedstock when either catalyst is used. Removal of more oil and gas and more complete pyrolysis are directly related to production of a better carbon black quality. For example, when the heating is stopped at the point where oil condensation initiates, the product from the non-catalyzed process contains in the range of 65% to 90% incompletely pyrolyzed rubber compared to on the order of 14% to 25% for the bentonite/metals catalyzed reaction.

15. FIGURE 2a shows a low magnification scanning electron microscopic image of carbon black that was processed at 1,200°F without catalyst. This material was derived experimentally to simulate carbon derived from high temperature pyrolysis systems. The figure shows thermal damage as characterized by pocked regions. In contrast, FIGURE 2b and 2c are low magnification scanning electron micrographs of carbon black that were produced in our process with a bentonite/metals or bentonite only catalyst formulations, respectively. These figures show the absence of damaged regions and smaller agglomerates than the non-catalyzed material.

16. High magnification scanning electron microscopic examination of samples from bentonite/metals catalyzed (FIGURE 3b) or bentonite only catalyzed (FIGURE

3c) vs. non-catalyzed (FIGURE 3a) reactions show that residue coating the carbon black is removed by application of either catalyst formulation. Use of the catalysts results in a carbon black having much less residue between the carbon particles.

17. FIGURES 4a, 4b, and 4c, are transmission electron microscope images (high magnification) that show the presence of deposits in the carbon black from a non-catalyzed run (FIGURE 4a) in contrast to a bentonite/metals (FIGURE 4b) or bentonite only (FIGURE 4c) catalyzed runs.

18. Poly-aromatic hydrocarbons were not detected by FTIR (Fourier Transform Infrared Spectroscopy) analyses in carbon black material derived from bentonite/metals catalyzed reactions.

19. FIGURE 5 summarizes the results of organic chemical analyses of carbon black samples contrasting bentonite/metals or bentonite catalyzed versus non-catalyzed runs. This graph shows that either catalyst decreases the organic contaminants (oil and pitch-like polymeric material) in pyrolysis carbon black. Also shown in FIGURE 5, is the hydrogen/carbon analysis, which demonstrates a marked decrease in relative hydrocarbon content in bentonite/metals or bentonite catalyst produced materials in contrast to the material from a non-catalyzed process, confirming the aforementioned organic analyses.

20. FIGURE 6 is a plot comparing sieve size for carbon black from bentonite/metals catalyzed, bentonite catalyzed and non-catalyzed reactions. It shows that carbon black product from either catalyzed reaction contains less fused material than the product from non-catalyzed reactions. This is indicative of the absence of organic contaminants (oil, pitch, incompletely pyrolyzed rubber), which act as a binder in the carbon black, producing large chunks.

21. We have also tested a continuous system that is capable of metering catalyst at a set rate and pyrolyzing 100 lbs to 1000 lbs of rubber. This system transfers the material through a 25 foot reaction tube under vacuum and has four electrically controllable temperature zones that are heated by infrared heaters. Temperature sensors throughout the reactor allow for control of reaction or reactor temperatures in each zone. Effects of varying catalyst formulation or system conditions on the reaction have been evaluated by comparing the energy (kilowatt-hours) required to maintain controlled temperatures.

22. Comparative 200 lb runs at zone temperatures of 650°F, 500°F, 750°F, and 790°F for zones 1, 2, 3, and 4, respectively, 0.32 ft/min material transfer rates through the reactor, and a metered catalyst feed rate of 1.1 weight percent relative to the rubber using 1 ¼ inch rubber chips with bentonite/metals, bentonite only or no catalyst in the continuous

reactor demonstrate significant differences between catalyzed and non-catalyzed reactions with respect to energy usage, product quality, and reaction progress. In particular, and as can be seen in FIGURE 7, decreased energy was required to maintain zone set temperatures with the use of catalysts (bentonite alone or bentonite/metal) as compared with no catalyst.

23. Additional observations were made from the comparative runs. For example, in runs using the bentonite/metals catalyst, the solid (carbon black) product appeared dry and friable and did not smell like oil, indicating a cleaner product. About 7.5 – 8.5 gallons of oil was produced and was readily condensed with no tars noted, indicating that more oil was removed from the carbon and that breakdown of the rubber was more complete than in the non-catalyzed reactions, which will be reflected in a cleaner carbon product. In addition, there is more oil generated. The flare was yellow to clear in appearance with no smoke or odor noted, which indicates a more saleable gas product and is good from an environmental standpoint. It is, also, indicative of more complete processing. The reaction was complete in 90 - 120 minutes, as noted by cessation of ignitable gas, and after completion no out-gassing or smoke was observed. On completion, the system was clean with no residues on auger or reactor walls.

24. When bentonite alone was used as a catalyst, the solid (carbon black) product appeared dry and friable with a trace level of incompletely pyrolyzed material. The material had a slight oil like smell. About 6.9 – 7.1 gallons of oil was produced and was readily condensed with minimal tars and greater than 0.6 gallons water noted. The flare was yellow to clear in appearance with some steam and little odor noted. The reaction was complete in 120 - 135 minutes, as noted by cessation of ignitable gas. After completion no out-gassing or smoke was observed, and the system was clean with no residues on auger or reactor walls. This indicates that, although the reaction was enhanced by this catalyst formulation, the reaction rate was slower and not as complete as the bentonite/metals formulation.

25. When no catalyst was used, the solid (carbon black) product contained chunks of un-pyrolyzed (rubbery, non-friable) material and was oil and pitch saturated, making it stick to the walls of the collection bin. In addition, it was smoky and had a harsh oil-like rubber odor. Less oil was produced (5 – 6 gallons). The heavy condenser oil tank line was plugged with pitch-like material. Out-gassing continued for five hours after completion of the run. This pitch buildup and out-gassing indicate incomplete breakdown of the rubber. During a major portion of the run time the flare exhibited a 4" to 6" smoke plumb below the ignitable gas. The flame was of typical height but, was smoky with an orange-yellow flame

and had a strong gassy sulfury odor. Upon extinguishing, it continued to produce a non-ignitable heavy white smoke that continued for 5 hours after shut down. This is indicative of poor quality gas and could pose environmental issues.

26. When no catalyst was used, examination of the reactor system showed the exhaust line to the heavy oil condenser to be plugged with pitch and the walls and auger were coated up to ¼ inch with oil, carbon and pitch. The reactor exit had up to a 10 inch buildup of oil and pitch saturated carbon with oil beads on embedded fiberglass fibers. Multiple cleanup cycles of catalyst spraying at increased levels followed by heat ramping followed by a standard pyrolysis run with an enriched catalyst was necessary to clean out the system fouling caused by several non-catalyst runs. From these tests we have demonstrated that pyrolysis of rubber in the absence of catalyst is ineffective at the temperatures and vacuum applied in our process. This is from an energy utilization, product quality, system maintenance and environmental contamination standpoint.

27. Based on our bench and pilot plant tests, we have documented that both catalyst formulations (bentonite alone and bentonite in combination with metals) contribute to the pyrolysis process in the following ways:

(i) They speed up the cracking of oil and polymeric material and allow the process to proceed at lower temperatures. This generates more oil, less pitch, and cleaner solid, liquid and gaseous products.

(ii) They allow the reaction to proceed with less energy input.

(iii) They promote movement of the material along the process path by preventing sticking.

(iv) They promote an exothermic reaction.

(v) They produce a cleaner product, which contains less oil and pitch, at lower temperatures and with less energy input. In fact, in the pilot reactor without one of the catalysts, the product is saturated with oil, pitch, and rubber and smokes during cool down.

(vi) They produce a lighter weight oil with less pitch, which is indicative of a more complete cracking reaction that is promoted by the catalysts.

(vii) They produce a cleaner burning gas as characterized by lack of smoke, soot, and odor and a transparent flame.

28. The substantial improvements noted above, all of which were provided by the use of a clay alone or clay/metals catalysts, could not have been predicted based on the cited references, or based on any other references known to us.

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

11-14-03

Date

Alan M. Levine

Alan M. Levine



## APPENDIX A PROFESSIONAL EXPERIENCE

### **Center For Hazardous Materials Research, Pittsburgh, PA Sept 1988 - Sept.1992**

Senior Research Scientist

- Headed a residue program consisting of HPLC, HPTLC, GC/ECD, GC/FID, and GC/MSD analyses of over 8,000 samples from eleven field sites.
  - Supervised analysis of oil in samples to monitor the effects of spills and to evaluate the effectiveness of remediation efforts.
  - Designed equipment to control chamber conditions in photolysis test cells.
- Director, Laboratory Programs/ Manager, Laboratory Programs
- Designed, implemented, staffed, and managed environmental fate laboratory programs in accordance with FIFRA and TSCA. The laboratory consisted of 15 employees operating in 12 rooms.
  - Studies were performed in strict compliance with GLP's.
  - Results of studies used by industrial sponsors to support chemical and pesticide registrations to the US EPA and corresponding foreign agencies.

### **Provac, Inc. (formerly Bactex, Inc.), Pittsburgh, PA Sept 1987 - Aug 1988**

Director of Manufacturing

- Participated in the formation of Provac as a spin-off corporation from Bactex, Inc.
- As a member of the management team, established and implemented company objectives.
- Supervised manufacturing scale-over and scale-up.
- Directed quality control assay refinement for a human urinary tract infection multivalent vaccine.

### **Bactex, Inc., Pittsburgh, PA Sept 1985 - Sept 1987**

Director of Manufacturing/ Manager of Manufacturing and Pilot Activities

- Designed, implemented, and managed a 7 laboratory complex for the manufacture of bacterial products under FDA/GMP compliance.
- Directed scientific staff in development, scale-up, and production of vaccines for clinical use.
- Designed a large scale facility to meet future needs.
- Prepared and managed annual budgets.
- Wrote and implemented SOP's.
- Directed HPLC fingerprinting and amino acid analysis.

### **Schering-Plough Corporation, Bloomfield, NJ Sept 1980 - Aug 1985**

Analytical Biochemist - Protein Chemistry Department

- Managed a GC/MS laboratory.
- Conducted research on GC/MS protein sequencing and HPLC systems.
- Developed software to predict fragmentation patterns of proteins and behavior of fragments.
- Set-up and debugged HPLC stations for R&D and in-process monitoring.

Process Biochemist

- Participated in design of a \$1M pilot plant facility for purification of genetically engineered interferon.
- Identified, purchased, and set-up production equipment for a pilot manufacturing facility.
- Performed research on bench-level and large-scale product isolation procedures.

### **The New York Blood Center, New York, NY Sept 1979 - Sept 1980**

Process Development Biochemist

- Setup and managed a pilot plant. Performed and directed development of large scale purification procedures for human blood products.

### **University of Pittsburgh, Department of Biological Sciences, Pittsburgh, PA Sept 1974 - Sept 1979**

Postdoctoral Fellow/ Thesis Research

- Developed and evaluated a vaccine against *P. aeruginosa*.
- Produced *Gonorrhea* and *Ecoli* vaccine antigens for clinical testing.
- Developed a software package for graphing data.

### **Stevens Institute of Technology, Hoboken, NJ Jun 1969 - Sept 1973 (Work Study)**

- Designed electronic, high vacuum, ultra-low temperature, and other laboratory equipment for a cryogenics research laboratory.

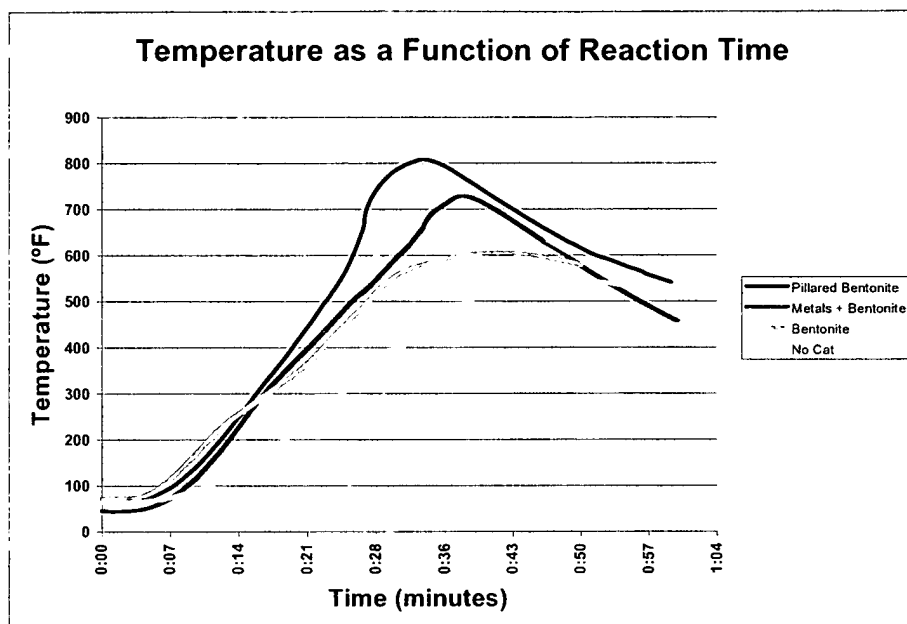
## APPENDIX B PUBLICATIONS

### PUBLICATIONS AND PRESENTATIONS

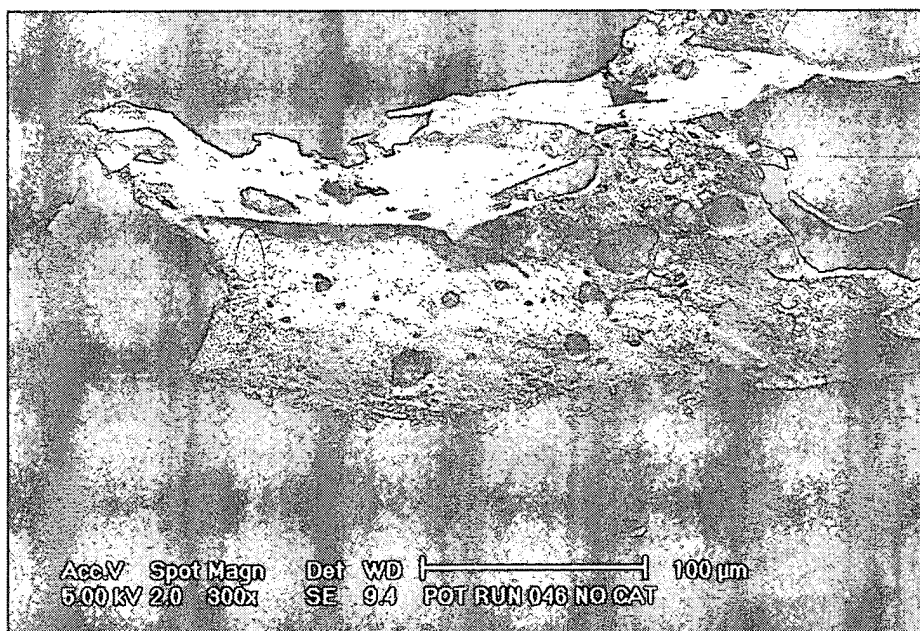
- Brinton, Charles; Bryan, James; Dillon, Jo-Anne; Guerina, Nicholas; Jacobson, Linda; Labik, Andrea; Lee, Simon; Levine, Alan; Lim, Sim; McMichael, John; Polen, Suzanne; Rodgers, Kenneth; To, Agnes C. -C.; To, Sam C. -M., Uses of Pili in Gonorrhea Control: I. The Role of Bacterial Pili in Disease; Purification and Properties of Gonococcal Pili and Progress in the Development of a Gonococcal Pilus Vaccine for Gonorrhea, Proceedings of the Symposium of Immunobiology of *Neisseria gonorrhoeae* by the American Society for Microbiology; held on January 18 - 20, 1978.
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- Levine, A. M., Studies on the Somatic Pili of *Pseudomonas aeruginosa* and their Role in Infectious Disease., Thesis- University of Pittsburgh Library, 1979.
- Silipigni, J. D.; Levine, A. M.; Brinton, C. C.; Sadoff, J.; Collins, H.; and Sidbry, H., Studies on the Antigenicity, Cross-Reactivity, and Efficacy of *Pseudomonas aeruginosa* Pilus Vaccines. Abstract B-18, American Society for Microbiology, 81st Annual Meeting held in Dallas, Texas, 1981.
- Nagabhushan, T. L.; Suprenant, H. P.; Le, H. V.; Kosecki, R.; Levine, A.; Reichert, B.; Sharma, B.; Tsai, H.; Trotta, P.; Bausch, J.; Foster, C.; Gruber, S.; Hoogerheld, J.; and Mercorelli, S., Characterization of Genetically Engineered Alpha-2 Interferon., Proceedings of the Interferon Workshop by the National Institute of Health, held on September, 28 - 30, 1983.
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- Casuccio, G. S.; Henderson, B.; Levine, A. M.; and Legg, W.; Electron Microscopic and Chemical Analyses of Environmental Samples Collected in the Middle East in Relation to Operation Desert Storm. In Preparation.
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- J Gallagher<sup>1</sup>, J Inmon<sup>1</sup>, S. Schlaegle<sup>2</sup>, A. Levine<sup>2</sup>, T Rogers<sup>3</sup>, J Scott<sup>1</sup>, R Sams<sup>1</sup>, F Green<sup>4</sup>, M Schenker<sup>5</sup>, K Pinkerton<sup>5</sup> 1NHEERL, US-EPA, RTP, NC, USA; 2RJ Lee Group Inc, Monroeville, Pa, USA; 3State U of NC, Raleigh, NC, USA; 4Dept. of Pathology and Laboratory Medicine, U of Calgary, Alberta, Canada; 5Dept of Epidemiology and Preventative Medicine, U of Ca, Davis, Ca; Tissue Remodeling in the Human Lung in Relation to Particle Concentration and Metal Content, USA Society of Toxicology Meeting in Nashville, Tennessee held in 2001 and Pittsburgh, Pennsylvania in March 31-April 4, 2003. J Gallagher<sup>1</sup>, J Inmon<sup>1</sup>, S Schlaegle<sup>2</sup>, A Levine<sup>2</sup>, T Rogers<sup>3</sup>, J Scott<sup>1</sup>, F Green<sup>4</sup>, M Schenker<sup>5</sup>, Norbert Menzel<sup>6</sup>, Klaus Whittmaker<sup>6</sup>, K Pinkerton<sup>5</sup> 1NHEERL, US-EPA, RTP, NC, USA; 2RJ Lee Group Inc, Monroeville, Pa, USA; 3State U of NC, Raleigh, NC, USA; 4Dept. of Pathology and Laboratory Medicine, U of Calgary, Alberta, Canada; 5Dept of Epidemiology and Preventative Medicine, U of Ca, Davis, Ca, USA, 6National Research Center for Environment and Health, Institute of Radiation, GSF; Germany Health Effects Indicators in Human Lung in Relation to Particle Concentration and Metal Content, Matter: Atmospheric Sciences, Exposure and the Fourth Colloquium on PM and Human Health, 2003.
- J Gallagher<sup>1</sup> (PI), A Levine<sup>2</sup>, K Wittmaack<sup>2</sup> (PI), J Inmon<sup>1</sup>, N Menzel<sup>2</sup>, K Pinkerton<sup>3</sup> 1US EPA, Human Studies Division, Epidemiology and Biomarkers Branch, Research Triangle Park, NC 27711, USA; 2 RJ Lee Group, Monroeville PA Inc. 3GSF National Research Center for Environment and Health, Institute of Radiation Protection, 85758 Neuherberg, Germany; 4Department of Epidemiology and Preventive Medicine, University of California, Davis, CA 95616, USA; Indicators of Health Effects Related to Particle and Metal Burden in Human Lung: Elemental Composition by Combined PIXE and ICP-AES Analysis, GSF EPA Inter-Laboratory Collaboration Presentation Series.
- Levine, A. and Tuminello, J.; Hazardous Waste Streams and Building Demolition; Detection, Handling, Disposal. Presented to the National Capital Chapter of the Environmental Information Association in Columbia,

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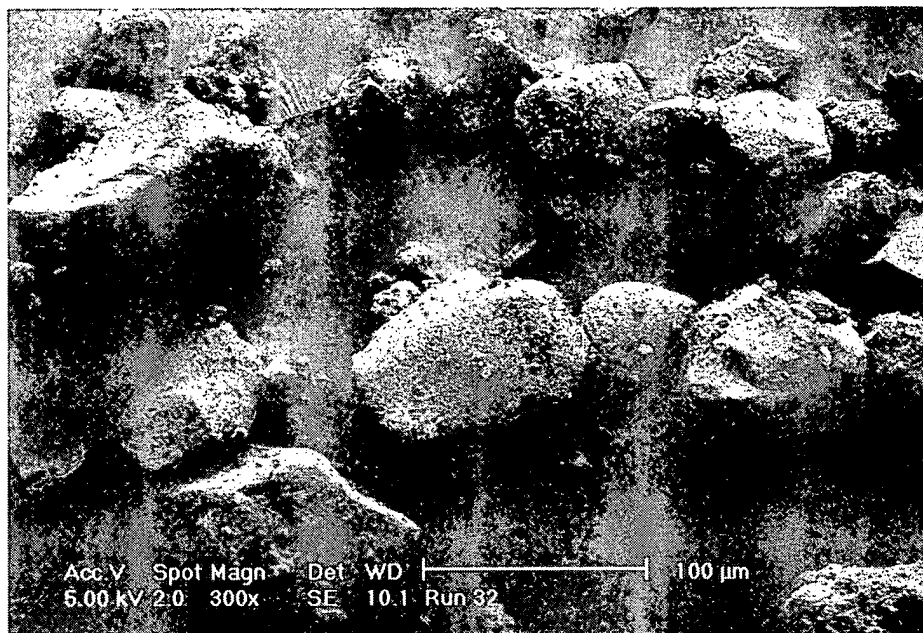
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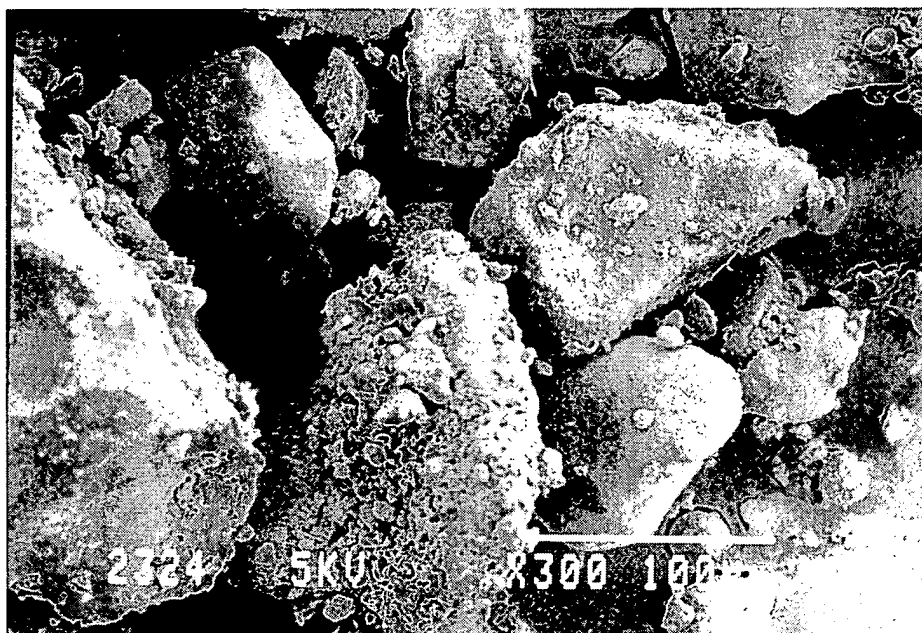
**FIGURE 1. Bench System Reaction Temperature at Equivalent Heat Inputs for Different Catalyst Formulations. Shows: Pillared Clay > Metals Plus Clay > Bentonite > Non-Catalyzed**



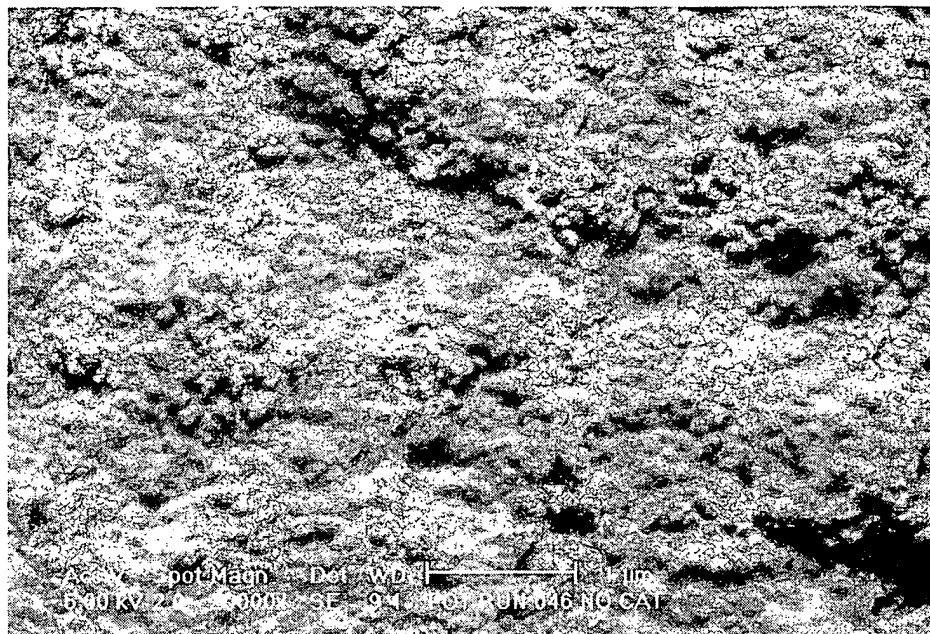
**FIGURE 2a. Scanning Electron Microscope Image of a High Temperature (1,200°F) Non-Catalyzed Pyrolyzed Carbon Black at 300x . Note the Damaged (Pocked) Regions.**



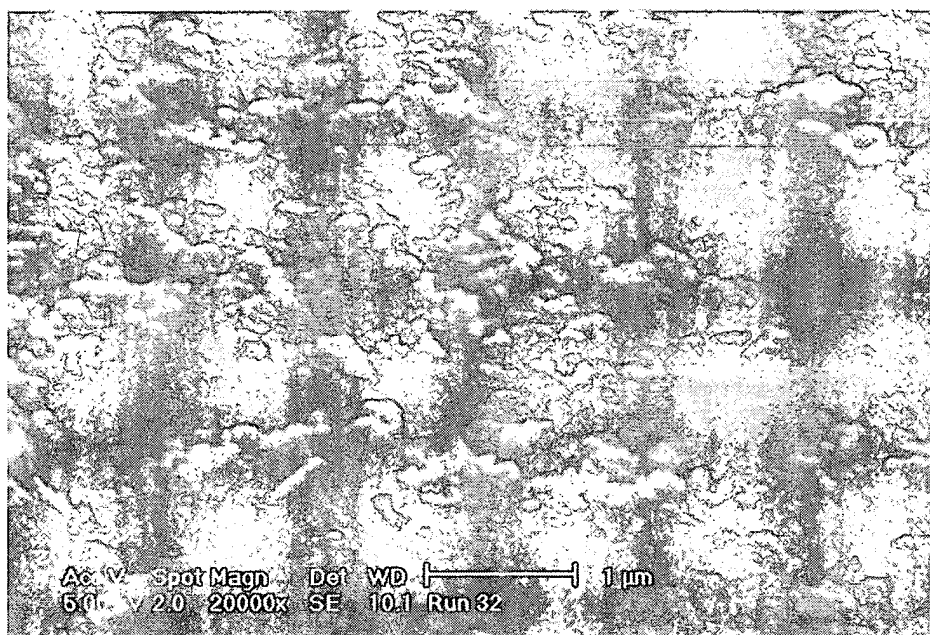
**FIGURE 2b. Scanning Electron Microscope Image of Carbon Black Produced by our Bentonite/Metals Catalyzed Pyrolysis Process at 300x. Note No Damaged (Pocked) Regions and Smaller Agglomerates.**



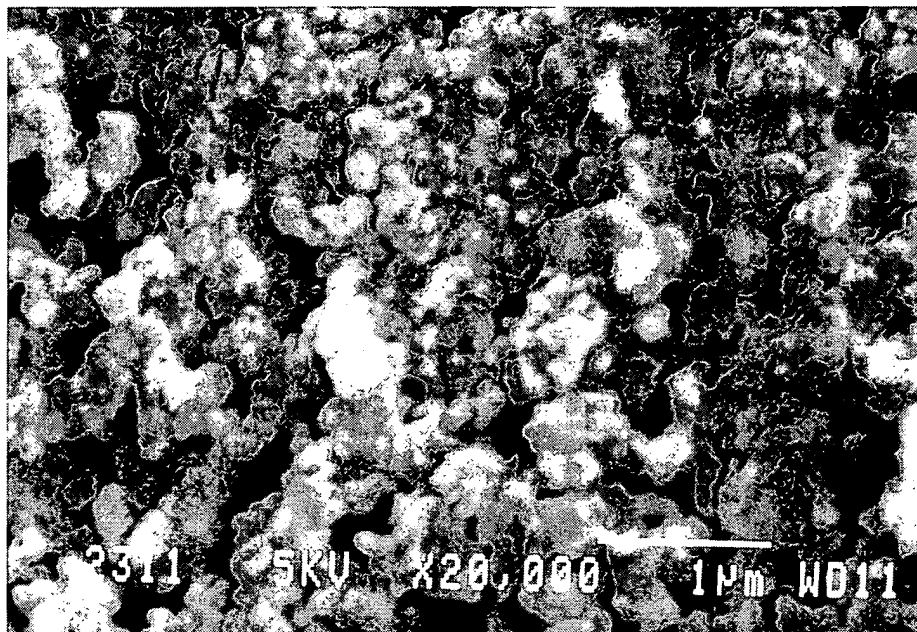
**FIGURE 2c. Scanning Electron Microscope Image of Carbon Black Produced by our Bentonite Only Catalyzed Pyrolysis Process at 300x. Note No Damaged (Pocked) Regions and Smaller Agglomerates.**



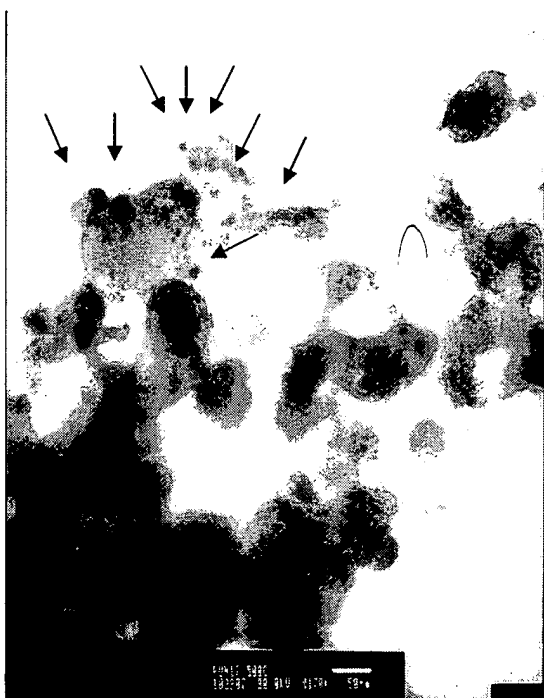
**FIGURE 3a. Scanning Electron Microscope Image of Carbon from a Non-Catalyzed Run at 20000x Magnification. Note the Residue Bridged between Spherical Carbon Black Particles.**



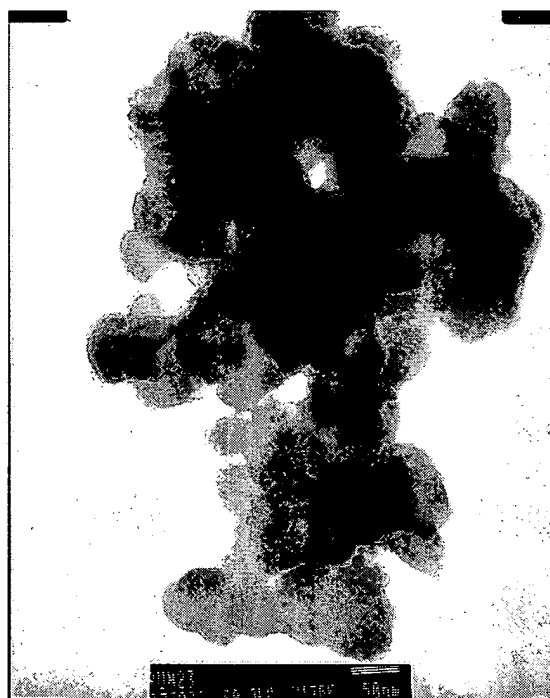
**FIGURE 3b. Scanning Electron Microscope Image of Carbon from a Bentonite/Metals Catalyzed Run at 20000x Magnification. Note the “Clean” Surfaces - No Residue between Spherical Carbon Black Particles.**



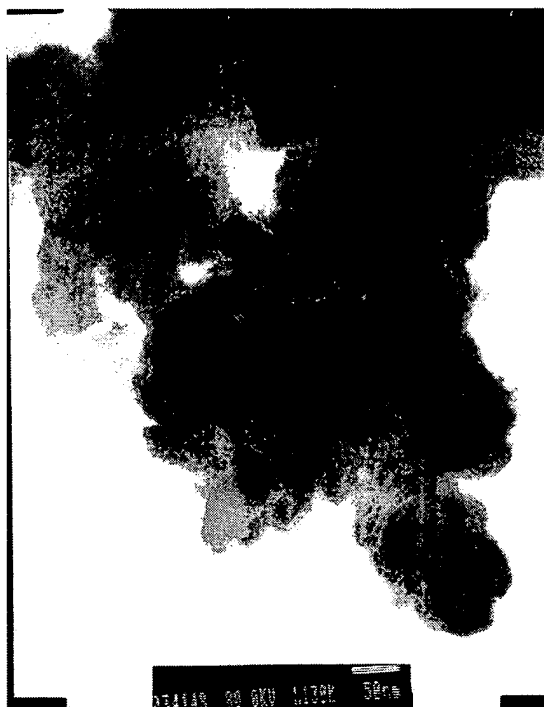
**FIGURE 3c. Scanning Electron Microscope Image of Carbon from a Bentonite Only Catalyzed Run at 20000x Magnification. Note the “Clean” Surfaces - No Residue between Spherical Carbon Black Particles.**



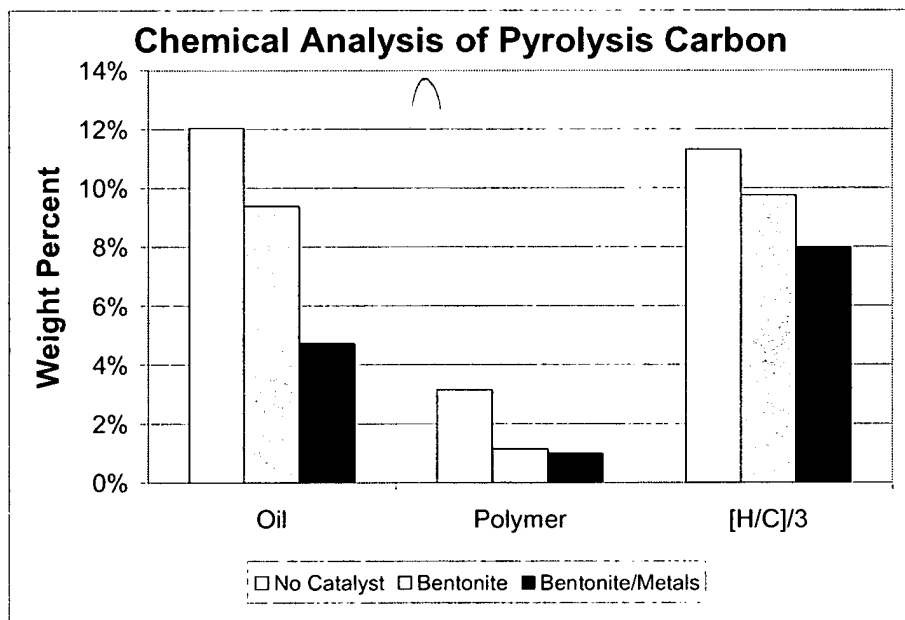
**FIGURE 4a. Transmission Electron Microscope (TEM) Image of Carbon Black from a Non-Catalyzed Run at 130,000 Magnification. Note the Dark Deposits.**



**FIGURE 4b. Transmission Electron Microscope (TEM) Image of Carbon Black from a Bentonite/Metals Catalyzed Run at 130,000x Magnification. Note the Absence of Dark Deposits.**

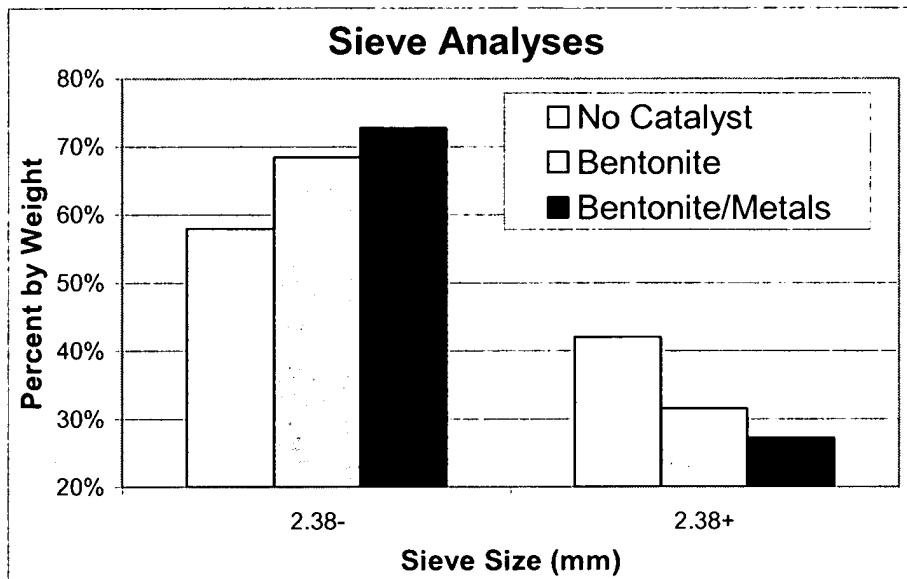


**FIGURE 4c. Transmission Electron Microscope (TEM) Image of Carbon Black from a Bentonite Only Catalyzed Run at 130,000x Magnification. Note the Absence of Dark Deposits.**

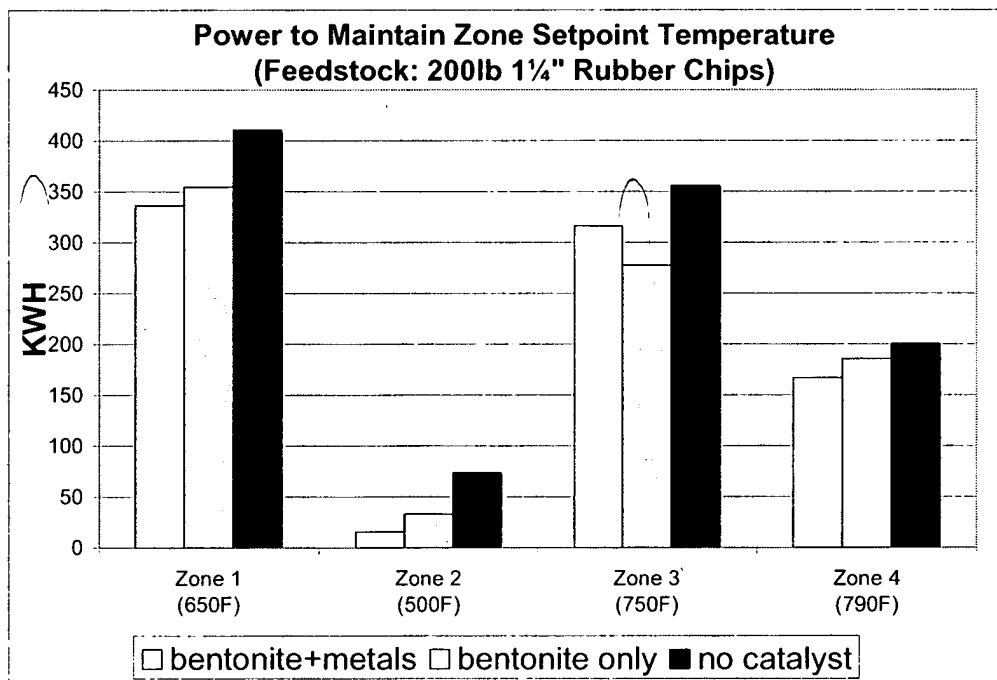


**FIGURE 5. Organic Analysis of Carbon Product from Catalyzed versus Non-Catalyzed Runs. Shows More Effective Removal of Organic Components When Catalyst is Used.**





**FIGURE 6. Comparison of Sieve Analysis of Carbon Black from Catalyzed Versus Non-Catalyzed Runs. Shows Less Material Stuck Together In Catalyzed Runs, Indicative of Less Polymer in the Material.**



**FIGURE 7. Power to Maintain Pilot Plant Zone Setpoint Temperature for Processing with Catalyst versus Without Catalyst**



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of	) Examiner: Thuan D. Dang
	)
RONALD E. NICHOLS et al.	) Group Art Unit 1764
	)
Serial No.: 09/925,391	) Entitled:
	)
Filed: August 9, 2001	) LOW ENERGY METHOD OF
	) PYROLYSIS OF HYDROCARBON
Atty Docket No.: 287122-00004-2	) MATERIALS SUCH AS RUBBER

**DECLARATION OF GARY A. COOKE**

I, Gary A. Cooke, being duly sworn hereby declare as follows:

1. I have a B.A. degree in geology from Case Western Reserve University, Cleveland, Ohio, awarded in 1973. I have a M.S. degree in geophysical sciences from the Georgia Institute of Technology, Atlanta, Georgia, awarded in 1977.

2. My employment history is as follows: From 2001 to present From 2001 to present, I have been a chemist at the U.S. Department of Energy Hanford 222-S Laboratory in Richland, WA. From 1995-2001 I was the president and CEO at Solutions International, Inc., Leechburg PA. I have been a lecturer from 1999 to present at Mercyhurst College, Erie PA, in the Department of Geology and Archaeology. I previously worked at RJ Lee Group, Inc., Monroeville PA, for the period of 1988-1995 as a research geochemist, as a manager of analytical chemistry, and as a senior research geochemist.

I worked at the University of Pittsburgh, Pittsburgh PA from 1983-1989 as an adjunct instructor in the Department of Geology and Planetary Sciences, and was a research associate and director of analytical services at the University of Pittsburgh's U-PARC Archaeometry Lab. I was a consulting scientist in materials analysis from 1983-1988. From 1977 to 1983 I worked at the Gulf Research and Development Company, Harmorville PA, as a senior geologist, from 1973-1977 at the Georgia Institute of Technology, Atlanta GA, as a research assistant, and at Case Western Reserve University as a laboratory technician from 1972-1973.

3. I am an author or coauthor of the publications and presentations listed in the attached Appendix A.

4. I have over 25 years experience in analytical chemistry, including familiarity with operation and maintenance of various analytical tools such as x-ray instrumentation, electron microscopes and microprobes, atomic absorption, emission and plasma arc spectroscopy, electrochemistry; visible, UV and IR spectrophotometry, and DTA/TGA/DSC. I have developed and taught analytical geochemistry courses to upper-class and graduate students at University of Pittsburgh, and I organized and managed several laboratories, including the 21 person analytical chemistry section at RJ Lee Group, Inc. I have also developed QA/QC plans and implemented QA/QC and certification for analytical chemistry laboratories.

5. In addition to my experience in analytical chemistry, I have over ten years experience, including extensive field experience, in petroleum geology and groundwater geochemistry, and have conducted fundamental research and development in aqueous geochemistry, surface chemistry, fluid flow and mineral diagenesis. I am knowledgeable in the chemistry and crystallography of clays, having specialized in the field of clay mineralogy as an undergraduate and graduate student, and I worked to develop and characterize synthetic clay catalysts at Gulf Research and Development Company.

6. I also have over ten years experience in materials analysis. I have conducted thousands of analyses on a wide variety of natural and manmade materials, to answer practical questions concerning toxic substances, constituent analysis, contaminant identification, failure analysis, process analysis, and other questions related to research and development.

7. I am not a named inventor on the present application. I have read the application, the recent office action, and the references cited by the Examiner, and in particular, the Gi reference, US Patent No. 4,463,203.

8. The Gi reference is cited as teaching the use of metals and clay as a catalyst in a method of pyrolysis. According to the Examiner, the metals in the bentonite used by Gi function in the same manner as the metal dust used in the present invention. The Examiner asserts that it would be obvious to use the amounts of metals presently claimed, as well as the ratio of metals to clay and the particles sizes of metals, as claimed, in view of the Gi reference. It is my well considered opinion that these assertions are incorrect.

9. The metals in the bentonite used by Gi do not function in a similar

manner to the metal dust used in the present invention for the following reasons:

(i) The metals found in bentonite are cations, that is they have a different valence state than elemental metals. For example, the metals Mg and Al have a +2 and +3 valence state, respectively, in the crystalline structure of montmorillonite, the principal component of bentonite. See, e.g., Newman, A. C. D. and Brown, G., 1987, *The Chemical Constitution of Clays*, Chapter 1 in: Chemistry of Clays and Clay Minerals, A. C. D. Neuman, ed. Mineralogical Society Monograph Number 6, John, Wiley and Sons, Inc. New York. Thus, the metals in clay are already oxidized.

(ii) The metals used in the present invention have a zero valence state, that is, they are in elemental form. Because they are in the zero valence state, they can undergo an oxidation reaction during the pyrolysis process and remove oxygen from the product.

(iii) The specific purpose of the addition of metals in the zero valence state is to provide a reagent that reacts with the organic material during pyrolysis in a manner that increases the yield of desirable hydrocarbons. When oxygen is removed by the oxidation of the metals, hydrogen gas is created. The hydrogen gas serves as a fuel, and is capable of hydrocracking the larger hydrocarbons released by the pyrolysis process, thus producing the desirable lighter hydrocarbons. Metals that are already oxidized, e.g. those found in clay, are not able to undergo this reaction. For all of the above reasons, it is my well considered opinion that the metals found in the bentonite of Gi do not function chemically as the metals presently claimed.

10. The purpose of the clay in Gi, as stated at column 3, lines 22-24, is to prevent the solid residue from sticking to the reactor. As shown above, the purpose of the addition of metals in the present invention is to provide a reagent that reacts with the organic material by undergoing the oxidation reaction. There is no teaching or suggestion in Gi that the clay can provide this function, and one skilled in the art would find no guidance in Gi as to how one could achieve this goal.

11. For all of the above reasons, it is my well considered opinion that Gi does not teach or suggest the use of metals in elemental form, nor does Gi teach the specific amounts of metals, the ratio of metals to clay, nor the particles sizes of the metals, as presently claimed in the present invention.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that

these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

11/6/2003

Date

Gary A. Cooke

Gary A. Cooke

## **ASSIGNMENT**

WHEREAS, we, **Ronald Nichols**, residing at 253 Normandy Road, Evergreen, Colorado 80439, **Alan M. Levine**, residing at 339 Coleman Drive, Monroeville, Pennsylvania 15146 and **Jules E. Langlois**, residing at 3732 Milligantown Road, New Kensington, Pennsylvania 15068, have invented certain new and useful improvements in the invention entitled **LOW ENERGY METHOD OF PYROLYSIS OF HYDROCARBON MATERIALS SUCH AS RUBBER** for which we filed Application Serial No. **09/925,391** having a filing date of **August 9, 2001**;

AND WHEREAS **RJ Lee Group, Inc.**, a Pennsylvania corporation, having its principal place of business at 350 Hochberg Road, Monroeville, Pennsylvania 15146, hereinafter called the "Assignee", is desirous of acquiring the entire right, title, and interest in and to said application and the inventions and improvements therein disclosed;

NOW, THEREFORE, in consideration of One (\$1.00) Dollar and other good and valuable consideration paid to us by said Assignee, receipt whereof we hereby acknowledge, we do hereby assign, sell, transfer, and set over unto said Assignee the entire right, title, and interest in and to said application and the inventions and improvements therein disclosed for the United States of America and all foreign countries and any Letters Patent which may issue therefor in the United States of America and all foreign countries and all divisions, reissues, continuations, renewals, and/or extensions thereof including all priority rights under the International Convention associated therewith for each country and the Union, said Assignee to have and to hold the interests herein assigned to the full ends of the terms of said Letters Patent and any and all divisions, reissues, continuations, renewals, and/or extensions thereof, respectively, as fully and entirely as the same could have been held and enjoyed by us had this assignment not been made.

The Commissioner of Patents and Trademarks is requested to issue such Letters Patent in accordance herewith. We covenant that we are the lawful owners of the said application, inventions, and improvements, that the same are unencumbered, that no license has been granted to make, use, or vend the said inventions or improvements or any of them, and that we have the full right to make this assignment.

And for the consideration aforesaid, we agree that we will communicate to said Assignee or the representatives thereof any facts known to us respecting said inventions and improvements, and will, upon request, but without expense to us, testify in any legal proceedings, sign all lawful papers, execute all divisional, reissue, continuation, renewal, and/or extension applications, make all rightful oaths, and generally do all other and further lawful acts, deemed necessary or expedient by said Assignee or by counsel for said Assignee, to assist or enable said Assignee to obtain and enforce full benefits from the rights and interests herein assigned. This assignment shall be binding upon our heirs, executors, administrators, and/or assigns, and shall inure to the benefit of the heirs, executors, administrators, successors, and/or assigns, as the case may be, of said Assignee.

EXECUTED 11/11/03

 (SEAL)  
RONALD NICHOLS

STATE OF COLORADO

COUNTY OF Tekkeron

)  
) SS: 557-68-8154  
)

Before me this 11<sup>th</sup> day of November, 2003, personally appeared **RONALD NICHOLS**, to me personally known to be the person who is described in and who executed the above instrument, and he acknowledged to me that he executed the same of his own free will for the purposes therein set forth.

  
Notary Public

(Affix Seal Here)

EXECUTED \_\_\_\_\_

\_\_\_\_\_  
**ALAN M. LEVINE** (SEAL)

STATE OF PENNSYLVANIA

)

) SS:

COUNTY OF ALLEGHENY

)

Before me this \_\_\_\_\_ day of \_\_\_\_\_, \_\_\_\_\_, personally appeared **ALAN M. LEVINE**, to me personally known to be the person who is described in and who executed the above instrument, and he acknowledged to me that he executed the same of his own free will for the purposes therein set forth.

\_\_\_\_\_  
Notary Public

*(Affix Seal Here)*

∩

∩



EXECUTED \_\_\_\_\_

(SEAL)

---

**JULES E. LANGLOIS**

STATE OF PENNSYLVANIA )  
 ) ss:  
COUNTY OF WESTMORELAND )

Before me this \_\_\_\_\_ day of \_\_\_\_\_, \_\_\_\_\_, personally appeared **JULES E. LANGLOIS**, to me personally known to be the person who is described in and who executed the above instrument, and he acknowledged to me that he executed the same of his own free will for the purposes therein set forth.

Notary Public

(Affix Seal Here)

## ***ASSIGNMENT***

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NOW, THEREFORE, in consideration of One (\$1.00) Dollar and other good and valuable consideration paid to us by said Assignee, receipt whereof we hereby acknowledge, we do hereby assign, sell, transfer, and set over unto said Assignee the entire right, title, and interest in and to said application and the inventions and improvements therein disclosed for the United States of America and all foreign countries and any Letters Patent which may issue therefor in the United States of America and all foreign countries and all divisions, reissues, continuations, renewals, and/or extensions thereof including all priority rights under the International Convention associated therewith for each country and the Union, said Assignee to have and to hold the interests herein assigned to the full ends of the terms of said Letters Patent and any and all divisions, reissues, continuations, renewals, and/or extensions thereof, respectively, as fully and entirely as the same could have been held and enjoyed by us had this assignment not been made.

The Commissioner of Patents and Trademarks is requested to issue such Letters Patent in accordance herewith. We covenant that we are the lawful owners of the said application, inventions, and improvements, that the same are unencumbered, that no license has been granted to make, use, or vend the said inventions or improvements or any of them, and that we have the full right to make this assignment.

And for the consideration aforesaid, we agree that we will communicate to said Assignee or the representatives thereof any facts known to us respecting said inventions and improvements, and will, upon request, but without expense to us, testify in any legal proceedings, sign all lawful papers, execute all divisional, reissue, continuation, renewal, and/or extension applications, make all rightful oaths, and generally do all other and further lawful acts, deemed necessary or expedient by said Assignee or by counsel for said Assignee, to assist or enable said Assignee to obtain and enforce full benefits from the rights and interests herein assigned. This assignment shall be binding upon our heirs, executors, administrators, and/or assigns, and shall inure to the benefit of the heirs, executors, administrators, successors, and/or assigns, as the case may be, of said Assignee.

EXECUTED \_\_\_\_\_

\_\_\_\_\_  
**RONALD NICHOLS** (SEAL)

STATE OF COLORADO )  
 ) SS:  
COUNTY OF \_\_\_\_\_ )

Before me this \_\_\_\_\_ day of \_\_\_\_\_, \_\_\_\_\_, personally appeared **RONALD NICHOLS**, to me personally known to be the person who is described in and who executed the above instrument, and he acknowledged to me that he executed the same of his own free will for the purposes therein set forth.

\_\_\_\_\_  
Notary Public

(Affix Seal Here)

EXECUTED \_\_\_\_\_

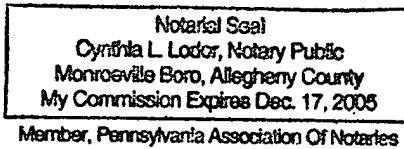
Alan M. Levine (SEAL)  
ALAN M. LEVINE

STATE OF PENNSYLVANIA )

) SS:

COUNTY OF ALLEGHENY )

Before me this 10<sup>th</sup> day of November, 2003 personally appeared  
ALAN M. LEVINE, to me personally known to be the person who is described in and who  
executed the above instrument, and he acknowledged to me that he executed the same of his own  
free will for the purposes therein set forth.



Cynthia L. Lodor  
Notary Public

(Affix Seal Here)

EXECUTED \_\_\_\_\_

\_\_\_\_\_  
**JULES E. LANGLOIS.** (SEAL)

STATE OF PENNSYLVANIA                    )  
  ) SS:  
COUNTY OF WESTMORELAND            )

Before me this \_\_\_\_\_ day of \_\_\_\_\_, \_\_\_\_\_, personally appeared  
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\_\_\_\_\_  
Notary Public

*(Affix Seal Here)*

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EXECUTED \_\_\_\_\_

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**RONALD NICHOLS** (SEAL)

STATE OF COLORADO )  
 ) SS:  
COUNTY OF \_\_\_\_\_ )

Before me this \_\_\_\_\_ day of \_\_\_\_\_, \_\_\_\_\_, personally appeared **RONALD NICHOLS**, to me personally known to be the person who is described in and who executed the above instrument, and he acknowledged to me that he executed the same of his own free will for the purposes therein set forth.

\_\_\_\_\_  
Notary Public

(Affix Seal Here)

EXECUTED \_\_\_\_\_

\_\_\_\_\_  
ALAN M. LEVINE (SEAL)

STATE OF PENNSYLVANIA

)

) SS:

COUNTY OF ALLEGHENY

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\_\_\_\_\_  
Notary Public

*(Affix Seal Here)*



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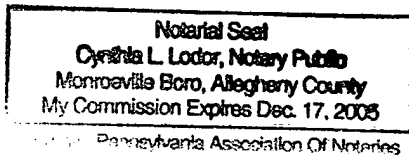
 (SEAL)  
**JULES E. LANGLOIS**

STATE OF PENNSYLVANIA

)  
) SS:  
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COUNTY OF WESTMORELAND

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Notary Public

(Affix Seal Here)